Modeling Dynamic Phase Changes in Zr—EOS and Kinetics

Carl Greeff and Sven Rudin (T-1), Frank Addessio and Eric Harstad (T-3), Paulo Rigg and Rob Hixson (DX-2), G.T. Gray, III (MST-8), Yusheng Zhao and Jian- zhong Zhang (LANSCE-12), and Marcus Knudson (Sandia National Laboratory); greeff@lanl.gov

Theoretical Division, in partnership with the Los Alamos National Laboratory's nuclear weapons programs, has historically been the home of advanced research in high-pressure equations of state (EOS) and shock physics. An important direction in this research at present is to improve our predictive capabilities for materials undergoing solid-solid phase changes. An interdivisional and multilaboratory collaboration is shedding new light on phase transitions elemental zirconium (Zr). Recent work has lead to new understanding of the phase diagram and EOS of this element and has revealed information on transformation kinetics and impurity effects. Ongoing work aims to improve the microscopic basis for the EOS and the description of the mechanical response.

A basic element of this work is the development of a high accuracy EOS, in the form of free energy functions for the individual phases. The resulting EOS is sufficiently accurate to allow us to make inferences about kinetics by comparing simulations with time-resolved data from dynamic compression experiments. Figure 1 shows the Hugoniot of Zr, which exhibits two distinct phase transitions. By correlating the Hugoniot with static compression and thermodynamic data, and electronic structure theory, we can associate the branches of the Hugoniot with definite crystal structures. The α-phase is hcp, the ω-phase is hexagonal with 3 atoms per unit cell, and β is bcc. The lower Hugoniot data lie along the metastable extension of the α-phase branch. This nonequilibrium behavior reflects the finite transformation rate.

A model for the kinetics of the α–ω phase transition has been proposed and applied to simulate dynamic experiments [1]. The model is successful in capturing the main features of wave profiles under various loading conditions. Figure 2 gives an illustration. The experimental VISAR profile was obtained in DX-2. The experiment consists of a sapphire impactor striking a Zr sample with a sapphire window. These investigations have shown a strong role of impurities in reducing the transformation rate. This is manifested in Fig. 2 through the higher transition threshold and broader wave in the lower purity sample. This same EOS and kinetic model was also successfully used to model isentropic compression experiments on the Sandia National Laboratory Z-machine.

Figure 1—
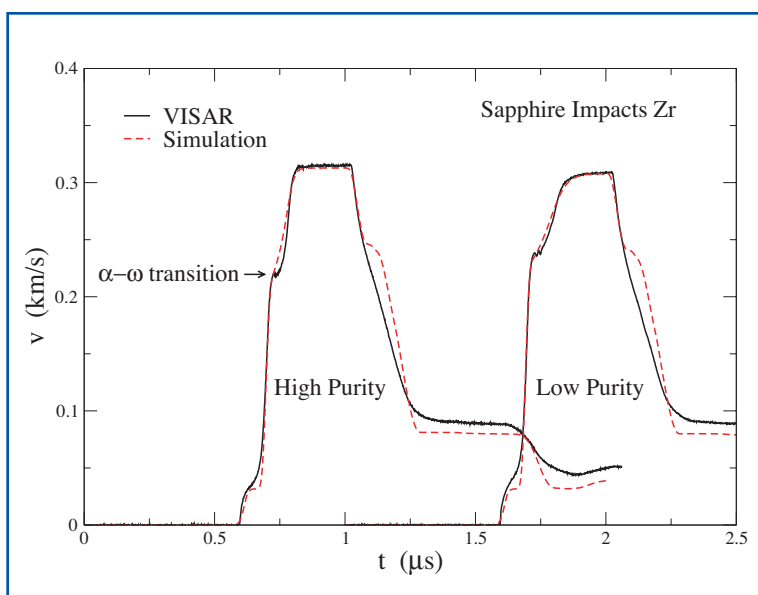*Hugoniot of Zr. Solid curve is equilibrium. Dashed curves are metastable extensions. Points are experimental data.*
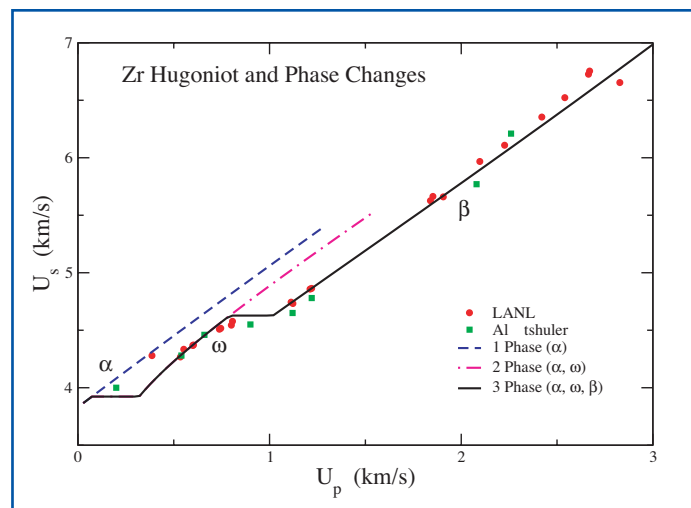
We are working to improve the EOS with lattice vibrational frequencies from electronic structure theory. These calculations tie into mechanical modeling by providing elastic moduli at high pressure, where they are difficult to measure. A complication that arises in Zr is significant anharmonicity. This is illustrated in Fig. 3, which shows the energy change associated with some high symmetry phonon displacements for Cu, Zr, and U. For a linear restoring force, the curves would be flat. In contrast to Cu, both Zr and U show substantial nonlinearity. Experimentally, this leads to strong temperature dependence of the corresponding frequency.

The shock-wave simulations discussed earlier in this paper use a simple elastic, perfectly plastic constitutive model, which leads, for example, to the difference with experiment in the shape of the release wave. Ongoing modeling efforts aim to improve the constitutive modeling and also to address the coupling of shear stress to the phase transition [2].

[1] C.W. Greeff, P.A. Rigg, M.D. Knudson, R.S. Hixson, and G.T. Gray, III, "Shock Compression of Condensed Matter – 2003," in *AIP Conference Proceedings* 706, M.D. Furnish and Y.M. Gupta, Eds. (AIP Conference Proceedings, Melville, NY, 2004).
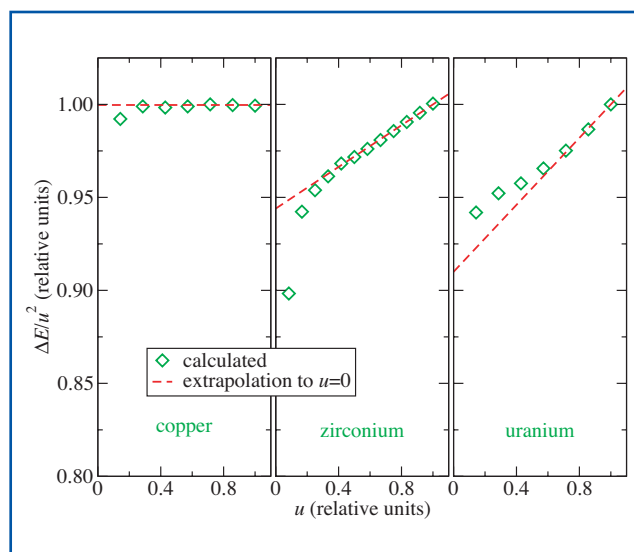
[2] E. Hartstad et al., "Modeling Phase Transformations with Strength in Zirconium," contribution on p. 156 in this volume.

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